Experimental Group on Rare Earth Compounds(Annual Report)

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Research Activities

(I) CRYSTAL GROWTH

1) Floating zone method (T. Komatsubara, S. Kunii and H. Shida)

Single crystal of Rare earth hexaboride (LaB_6, CeB_6, PrB_6, NdB_6) are obtained by floating zone method. Growth temperature is about 2500°C and obtained single crystal has 10 mm diameter 40 mm length in dimension.

2) Al flux method (M. Kasaya)

Single crystal of Rare earth hexaborides (EuB_6, SmB_6) are obtained by Al flux method. The Grouth of temperature of this method is about 1400°C and fairly lower than floating method. Therefore, this method has a advantage for avoiding the creation of vacancy occuring in high temperature crystal growth.

3) Bridgman method (T. Suzuki, M. Sera, A. Ochiai and A. Kamijo)

Single crystal of Rare earth chalchogenide (EuTe, Sm_3Se_4, Sm_3Te_4, SmTe, $Gd_{3-x}\square_xS_4$) are obtained by Bridgman method in the fused W and Mo crucible. The vapor pressure of these compound are very high in melting temperature ∿2500°C. So it must be used in close system of W and Mo crucible in crystal growth.

(II) MAGNETIC SEMICONDUCTOR SYSTEMS

A) Narrow Gap Semiconductor

1) Influence of Conduction Electron in a Heavily-Doped Magnetic Semiconductor, A Eu^{151} Mössbauer Study of Europium Hexaboride (M. Kasaya)

The europium is divalent and no evidence of mixed valence is found in any of five different samples of europium hexaboride. The magnetic order evolves from ferromagnetic in near-stoichiometric material to antiferromagnetic in $EuB_{5.8}C_{0.2}$ passing through a mictomagnetic region with inhomogeneous exchange around $EuB_{5.9}C_{0.1}$. There it is suggested that ferromagnetic

europium clusters around the carbon donors are coupled antiferromagnetically by direct exchange of electrons from adjacent donors.

2) Electronic Structure of EuB_6, Transport and Magnetic Properties (T. Kasuya, M. Kasaya and Y. Isikawa)

In non-magnetic $R^{++}B_6$, it is shown that the most common defect R^{++} vacancy, can trap third hole creating a compensation conduction electron due to the narrow gap semiconductor character. The latter is trapped loosely and thus easily causes n-type character. Various transport properties are explained on this model. In magnetic EuB_6, the above defect center makes a giant moment cluster. Furthermore, due to the antibonding effect with the 4f levels, the valence bands of the parallel spin are pushed up into the conduction bands in ferromagnetic state creating many conduction electrons. Various anomalous magnetic and magneto transport properties are explained on this model.

2) ESR of EuB_6 (S. Kunii and T. Kasuya)

Electron spin resonance (ESR) in antiferromagnetic EuB_6 single crystal was observed and compared with the results of ESR in EuB_6 ferromagnetic bulk poly-crystals. No anisotropy was observed in EuB_6 single crystal in all temperatures and all frequency bands in this experiment. Resonance points in single crystal move rapidly to lower fields as temperature is lowered. The shifts of the resonance field should be interpreted by a dynamical anisotropy field. Resonance line width increases with decreasing temperature especially for the cases of single crystal and lower frequencies.

4) Effects of Carbon on the Magnetism of EuB_6 (M. Kasaya)

Magnetization has been measured as function of temperature and magnetic field for compounds of composition $EuB_{6-x}C_x$ ($0 \leq x \leq 0.21$). It is suggested that samples with x=0 and 0.21 are ferromagnetic and antiferromagnetic, respectively, whereas those with intermediate composition are mictomagnets.

B) Impurity State and Randomness in Magnetic Semiconductor

1) Transport and Magnetic Properties of $Gd_{3-x}\square_xS_4$ (A. Kamijo, A. Takase, Y. Isikawa, S. Kunii, T. Suzuki and T. Kasuya)

Magnetic and transport properties of $Gd_{3-x}\square_xS_4$ were measured. Magnetic properties are understood by a Homogeneous d-f exchange model with a small Fermi energy treated before by us. Transport properties are not understood by a usual separated bound magnetic polaron model. Magnetic Wigner localization is proposed.

2) Magnetic Impurity State in EuTe (M. Sera, A. Takase, M. Kawakami, T. Suzuki and T. Kasuya)

Magnetic and Transport properties of a single crystal EuTe with $n_c = 1.7\times10^{18}$ cm^{-3} were measured. They behave completely differently from those reported by MIT group before. The results including MIT samples are interpreted on the different kinds of impurity state in which amorphous magnetic

Wigner crystal localization plays important role.

(III) DENSE KONDO SYSTEMS

1) Magnetoelastics and Ultrasonic Absorption in CeB_6 (S. Kunii, T. Komatsubara and T. Kasuya)

Both thermal expansion and magnetostriction show unusual anisotropic behavior even in the paramagnetic, or dense Kondo, region. In the ordered region, both magnetostriction and ultrasonic absorption show rich of structure. A model is proposed.

2) Magnetic and Electronic properties of CeB_6 (T. Komatsubara, T. Suzuki, M. Kawakami, S. Kunii and T. Kasuya)

Detailed magnetic, transport, specific heat and optical measurements were done on single crystals of CeB_6 down to 1.25 K and up to 400 kOe of applied field. Various unusual properties are understood by the cooperative phenomena of the dense Kondo state on the doublet and the anisotropic exchange interaction among the quartets.

3) Raman Scattering in CeB_6 (S. Kunii, T. Komatsubara and T. Kasuya)

Raman spectra of phonons in CeB_6 were measured. The observed three peaks were assigned to the A_{1g}, E_g and F_{2g} phonon modes. The frequencies of the modes suggest that the Ce ion is in the trivalent state.

(IV) VALENCE FLUCTUATING SYSTEMS

1) Valence Fluctuation in Sm_3Se_4 (A. Ochiai, T. Suzuki and T. Kasuya)

A single crystal of Sm_3Se_4 was prepared and the susceptibility was measured carefully. Simple analysis gives similar result to that of Sm_3S_4 by Wachter. Analysis by the IBM model including exchange interactions correctly also fits the data with reasonable values of exchange constants.

2) Transport and Magnetic Properties of SmB_6 (M. Kasaya, Y. Isikawa and H. Kimura)

Lattice parameter, magnetic susceptibility and L_{III} absorption measurements have been made on $Sm_{1-x}La_x$ and $Sm_{1-y}Yb_yB_6$ alloys. The results indicate that La^{3+} substitution for Sm in SmB_6 decreases the Sm valence, whereas substitution with Yb^{2+} increases the Sm valence. It is found that the contribution of Sm^{3+} to the susceptibility disappears for a wide range of composition. For the measurements of transport properties, we have prepared single crystals by the Al-flux method. Both La and Yb substitution decrease the resistivity at low temperature and change the sign of Hall Effect.

(V) MAGNETIC INTERACTION

1) Electron spin resonance of EuSe (H. Fukuma and S. Kunii)

Ferromagnetic and Ferrimagnetic resonance of EuSe were measured to study of the magnetic anisotropy. The Experimental result show a quite different

from those of usual magnetic material due to several computing magnetic interaction of EuSe. (Exchange interaction of ferro and Antiferro, dipole interaction, biquadratic spin exchange interaction) The technique to obtain very small sphere of 70μ diameter is established to measure FMR of EuSe.

2) Forbidden Transitions of Mn^{2+} in Zinc-Telluride Induced by Hyperfine Interactions (S. Kunii)

The radio frequency field parallel to the static magnetic field induces a simultaneous flipping of an electron spin and a nuclear spin of Mn^{2+} in zinc-telluride. The positions and the intensity of these transitions are provided by hyperfine interactions.

3) Hyperfine Interaction in Pr_3Tl (M. Kawakami and T. Kasuya)

The specific heat C_p of the ordered Pr_3Tl has been measured between .4 and 4 K. It is expressed as $C_p = 0.25T+0.530T^{-2}$ J/mole K at low temperatures. The second term gives the square average of magnetization $\langle M^2\rangle^{1/2}=1.3\mu_B$/Pr. The spontaneous magnetization $\langle M\rangle$ of the same sample has also been found to be $.75-.8\mu_B$/Pr. Pr_3Tl is thought to have a canting type ordering with the common ferromagnetic direction on the [111] direction to the [100] direction.

(VI) 3d TRANSITION METAL COMPOUND

1) NMR Study of Ferromagnetism in $V(Ir_{1-x}Pt_x)_3$ (M. Kawakami)

The zero-field NMR signal in magnetic $V(Ir_{1-x}Pt_x)_3$ with the Cu_3Au-type structure has been observed in the frequency range of 10-80 MHz by spin echo method at 1.7 K. The peak of the spectrum which is attributable to the ^{51}V nucleus has a nearly constant frequency -55MHz for $x>0.69$, corresponding to about -50 kOe. The dependence of the signal intensity on x is interpreted. VPt_3 is thought to have a magnetic ordering below 206 K, at which the electrical resistivity has an anomaly.

2) Temperature Dependences of Magnetization and Spin Waves in MnP (A. Takase and T. Kasuya)

The temperature dependence of the easy axis magnetization in the ferromagnetic phase of MnP is measured to an accuracy of $\Delta M/M<\pm 5\times 10^{-4}$ over a range from 4.2 K to 280 K. Instead of the usual $T^{3/2}$ law, TlnT dependence holds up to about $T_c/2$. The following spin wave dispersion is expected to exsist at 0 K, along the a axis it is nearly constant up to $q_a=Q$ and increases very rapidly beyond Q, where Q is the screw propagation vector, while for perpendicular to the a axis it is quadratic in Q . At a finite temperature, the calculated two-magnon renormalization causes increase of the magnon energy near $q_a \sim Q$ which induces an anomalous suppression for decrease of magnetization in MnP. The ferro-screw phase transition seems also to be due to this effect.

3) Low Temperature Specific Heat of MnP: Experiment and Analysis (A. Takase and T. Kasuya)

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3) Magnetic impurity state in EuTe, M. Sera, A. Takase, M. Kawakami, T. Suzuki and T. Kasuya, ibid.
4) Effects of carbon on the Magnetism of EuB_6, M. Kasaya, J.M. Tarascon, J. Etourneau, P. Hagenmuller and J.M.D. Coey, J. de Physique C-5(1979) 393.
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13) NMR study of ferromagnetism in $V(Ir_{1-x}Pt_x)_3$, M. Kawakami and T. Goto, J. Phys. Soc. Japan 46(1979),1492.
14) Temperature dependences of magnetization and spin waves in MnP, A.Takase and T. Kasuya, J. Phys. Soc. Japan 47(1979),491.
15) Low temperature specific heat of MnP, Experiment and analysis, A. Takase, H. Yashima and T. Kasuya, J. Phys. Soc. Japan 47(1979),531.

Master Thesis (March 1980)

1) Transport and Magnetic Properties of $Gd_{3-x}\square_xS_4$, Atsushi Kamijo.
2) Valence Fluctuation in Sm_3Se_4, Akira Ochiai.